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ATOMIC PHYSICS IN STRONG FIELDS

FINAL REPORT

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— Final Report —

I. Introduction

In this final report we present a summary of recent research accomplishments under the DOE support (FG02/90ER14159). In this grant period (FY90–97), some 36 papers have been published. [See Part IV for a list of publications.] Significant progress has been made in several areas, both in the development of new theoretical and computational methods, their applications to experimental observations, and in the prediction of some novel new high-intensity phenomena. More detailed discussions on individual research project can be found in previous DOE annual reports and in published articles.

II. Summary of Recent Research Accomplishments

A. *Time Evolution and Multiphoton Ionization of Rydberg Wavepacket in Microwave Fields*

It has been demonstrated experimentally that it is now feasible to create a Rydberg wavepacket from the ground state by a short pulse laser excitation. The Rydberg wavepacket so generated can be localized in radial space and will mimic the classical radial motion of a charged particle in a Coulomb field. As such the study of the dynamics of Rydberg wavepacket provides a unique opportunity, both theoretically and experimentally, for probing the correspondence between classical and quantum mechanics.

Recently we have initiated a theoretical study of the time evolution of Rydberg *wavepacket*, initially localized in radial space and mimicking the classical particle, undergoing *microwave multiphoton ionization*. The dynamical behavior of the Rydberg *wavepacket* is found to be different from that of Rydberg atoms initially prepared at given Rydberg *eigenstates*. We found that the Rydberg wavepacket can exhibit collapse and revival behavior in the short time regime. Further, both the microwave ionization rates and the quantum phase space trajectories of the Rydberg wavepackets exhibit qualitatively different behavior in the small scaled-frequency ($\omega_s (\equiv \omega n_0^3) \ll 1.0$) and large scaled-frequency ($\omega_s \geq 1.0$) regimes.

B. *Multiphoton Detachment of H^-*

The study of multiphoton detachment of H^- is a subject of much current interest, stimulated

mainly by the recent experiments at LAMPF in Los Alamos. The experimental setup uses a relativistic Doppler effect, allowing the continuous tuning of the laser frequency (in the center of mass or atom frame) over a wide range of photon energies.

For moderately strong laser intensities (10^9 to 10^{11} W/cm²) used in the experiments, doubly excited states are far above the ionization threshold of the ground state and can be safely ignored. We thus first construct an accurate one-electron model potential which reproduces precisely the known H⁻ binding energy and the low-energy e-H(1s) elastic scattering phase shifts. The model potential so constructed gives rise to essentially exactly the one-photon detachment cross sections obtained from previous *ab initio* two-electron correlated calculations, demonstrating its adequacy.

We then first performed *perturbative* calculations of multiphoton detachment of H⁻. Generalized n-photon detachment cross sections, in both linearly and circularly polarized fields, are evaluated based on the lowest nonvanishing order perturbation theory. An accurate and efficient numerical procedure is developed for the solution of the set of coupled inhomogeneous differential equations using the Dalgarno-Lewis' procedure for implicit summation of intermediate states. Two- to eight-photon detachment cross sections are determined for a wide range of photon energies. Comparison with several existing *ab initio* two-electron calculations (two- and three-photon detachment cross sections) is made. It appears that the present study provides the first consistent results with the accuracy of higher-order detachment cross sections ($n > 3$) comparable to those of lower orders ($n = 2, 3$). These weak-field perturbative results are then used to simulate the experimental intensity-averaged multiphoton detachment rates. The comparison with the experimental data is shown in Figure 1. While the theoretical results are in reasonable good agreement with the experimental data (which has an error bar of a factor of five), the perturbative calculations tend to predict a sharper transition nearby each n-photon threshold, suggesting that *nonperturbative* treatment may be needed. [Note that while the intensity range considered here is not very large in the absolute scale, the H⁻ ion can still be subject to strong field effects since it possesses a small binding energy.]

We proceed next to perform a series of *nonperturbative* studies of intensity- and frequency-dependent multiphoton detachment of H⁻ in monochromatic fields using the non-Hermitian Floquet formalism. A new computational procedure, the *complex-scaling generalized pseudospectral* (CSGPS) method is introduced for the discretization and solution of the time-independent non-Hermitian Floquet Hamiltonian. The CSGPS method does not require the calculation of the potential

matrix elements and is computationally more advantageous than the traditional basis set expansion-variational methods. [A more detailed discussion of this method is given in the section of the description for proposed research and in references 2 and 5.] We have determined the complex quasi-energies ($E_R - \Gamma/2$), of the ground state of H^- for the laser intensity range (1 to 40 GW/cm²) and the frequency range (0.20 to 0.42 eV), providing directly the intensity- and frequency- dependent a.c. Stark shifts and total multiphoton detachment rates. Some typical results are shown in Figure 2. These detailed results are then used to simulate the experimental intensity-averaged multiphoton detachment rates, taking into account the experimental conditions for H^- and laser beams. The theoretical results (without any free parameters) are in very good agreement with the experimental data (well within the experimental error bar) both in the absolute magnitude and in the threshold behavior (see Fig. 3).

In the presence of strong fields, multiphoton and above threshold ionization (MPI/ATI) can occur. In this case, the information on the angular distribution and partial rates are essential. We then proceed to develop a method for the determination of these quantities based on a formal analysis of the integral form of the Schrödinger equation. Further, we introduce a "back rotation" procedure for the analytical continuation of the complex-scaled quasienergy wavefunctions back to the real axis, allowing the calculation of the electron energy and angular distributions and partial rates in an accurate and more straightforward manner. The complex-scaled quasi-energy wavefunctions can be determined by the non-Hermitian Floquet formalism and the CSGPS method. Alternatively, a nonperturbative *adiabatic* Floquet theory has been formulated, which is valid when the laser frequency is smaller than the binding energy of the negative ions, providing an efficient new approach for the study of MPI/ATI of negative ions such as H^- . Figures 4(a)–4(c) show the ATI spectra of H^- at 10.6 μm for three different laser intensities and Figure 5 shows an example of the angular distributions from different electron peaks. Table I shows the results of converged ATI partial rate calculations.

More recently we have performed a non-perturbative Floquet calculation of the electron angular distributions and partial widths for multiphoton above-threshold detachment of H^- by 1064 nm radiation. When a beam of H^- ions is used as a target, the angular distributions in the frame where the ions are at rest can be extracted from the energy (angle-integrated) distributions in the laboratory frame. Our study of this process was stimulated by the recent experiments in Los Alamos measuring the electron angular distributions associated with two-photon detachment of H^- . The

calculations were performed for the linearly polarized laser field with the wavelength 1064 nm and several intensities in the range 10^9 – 10^{12} W/cm². With the increase of the number of photons absorbed above the threshold, the angular distributions become more stretched along the field direction manifesting, however, a few additional maxima. Besides the numerical data for the angular distributions we present also the coefficients of expansion on the basis of Legendre polynomials. Our results show that the detached electrons exit with primarily a D wave for the two-photon excess photon detachment process. This prediction has been confirmed by the recent Los Alamos experiment.

C. *Multiphoton and Above-threshold Ionization in Two-Color Fields*

The works described above are for one-color fields. Recently there are also considerable interest in the study of MPI/ATI processes in two-color fields. We have recently presented a general theory of electron energy and angular distributions and partial rates in two-color (two-frequency) fields for both commensurate and incommensurate frequencies based on the extension of the many-mode Floquet theorem and the integral form of the Schrödinger equation. The theory is applied to the first study of multiphoton above-threshold detachment of H⁻ for the fundamental frequency of CO₂ laser (wavelength 10.6 μ m) and its third harmonic. The calculations were performed for the fundamental field intensity 10^{10} W/cm² and harmonic field intensities 10^8 and 10^9 W/cm². The results show some novel features. First, both the total and partial rates and the angular distribution show a strong dependence on the relative phase between the two fields. The total rate is the largest for the phase $\delta = 0$, and the smallest for $\delta = \pi$. Further, a small admixture of harmonic field can lead to either enhancement or decrement of the H⁻ detachment rates, depending upon the relative field strength of the harmonic to the fundamental fields. Figures 6 and 7 show some typical results of the phase-dependent ATI electron energy and angular distributions in two-color laser fields.

D. *Multiphoton Above-Threshold Ionization by Intense Laser Pulses—A New Adiabatic Approach*

We have developed a new *adiabatic* approach for nonperturbative treatment of multiphoton above-threshold ionization by intense smooth laser *pulses*. We show that the energy and angular distributions of emitted electrons in the laser *pulses* can be expressed via the photodetachment amplitudes in the *monochromatic* laser fields:

$$\frac{dP(\mathbf{k})}{dE d\Omega} = \frac{k}{(2\pi)^3} \left| \sum_n \int_{-\infty}^{\infty} dt \exp[iE(\mathbf{k})t + i(2\omega)^{-2} \int_{-\infty}^t F^2(\tau) d\tau - i \int_{-\infty}^t \Delta \epsilon(\tau) d\tau - i(E^{(0)} + n\omega)t] A_n(\mathbf{k}, t) \right|^2,$$

where $\mathbf{k}$ is the electron momentum, $E^{(0)}$ is the unperturbed energy of the initial state and $\Delta \epsilon(t)$ is a complex value including the instantaneous AC Stark shift and (minus) halfwidth of the state. The monochromatic amplitudes $A_n(\mathbf{k}, t)$, depending on time via the field envelope $F(t)$, can be determined by means of the *adiabatic* Floquet theory. In practice, a pulse containing tens of field oscillations is sufficient for the use of the theory. The longer the pulse, the more exact the theory. Another approximation is the use of only one adiabatic quasienergy state for the description of electron wave function. This is well justified for negative ions, such as H^- , since there is only one bound state. We apply the theory to the study of the pulse shape effects on the electron energy and angular distributions for the multiphoton above-threshold detachment of H^- at 10.6 μm . Figure 8 shows an example of the angle-integrated electron energy spectra dP/dE for multiphoton above-threshold detachment of H^- in a Gaussian pulse with laser peak intensity $5 \times 10^{10} \text{ W/cm}^2$ and pulse length T : (A) $T = 5 \text{ ps}$, (B) $T = 10 \text{ ps}$, and (C) $T = 20 \text{ ps}$. [(D) is the monochromatic result for the same peak intensity.] The oscillatory satellite structures to each main ATI peak are due to the interference of the electrons detached on the raising and falling edges of the pulse. Simple analytical formulas describing these subpeak structures are also presented. The method can be extended to the general treatment of multiphoton above-threshold detachment of negative ions by arbitrarily shaped (smooth) laser pulses.

E. Laser-Induced Chemical Bond Softening and Hardening of H_2^+ and D_2^+ in Intense Laser Fields

Recent advances in high-intensity experiments on multiphoton and above-threshold ionization (MPI/ATI) of atoms have stimulated growing interest in the study of multiphoton and above-threshold dissociation (MPD/ATD) dynamics of diatomic molecules. In the presence of external laser fields, all the vibrational levels of H_2^+ molecules in the ground ($1s\sigma_g$) state become coupled to the dissociative continuum of the upper (repulsive) electronic state $2p\sigma_u$ (see Fig. 9). As a result, all the vibrational levels now turn to (shifted and broadened) quasi-bound states, which we shall call the vibrational quasienergy (VQE) resonances below. We have performed a series of theoretical studies of MPD/ATD of H_2^+ and D_2^+ in intense monochromatic and pulsed laser

fields. These include: (a) the development of a complex-scaling Fourier grid Hamiltonian (CSFGH) method for the discretization and solution of the time-independent non-Hermitian Floquet Hamiltonian associated with MPD/ATD processes in monochromatic fields. The method is simple to implement and allows efficient and accurate determination of the complex energies ($E_R, -\Gamma/2$) of both low-lying and highly excited vibrational quasi-energy (VQE) resonance states simultaneously, using only a small number of grid points (~ 80). The real parts (E_R) of the complex VQE's give rise to the a.c. Stark-shifted vibrational energies and the imaginary parts (Γ) provide directly the total MPD/ATD rates. (b) The prediction of a novel new high-intensity molecular phenomenon: molecules (initially prepared at high-lying vibrational levels) can become more stabilized in stronger laser fields, i.e. the lifetimes (MPD rates) of these high-lying VQE resonances increase (decrease) with increasing laser intensity and the chemical bond is harder to break at higher fields, a phenomenon may be termed "bond hardening." (See Fig. 10). [The highest laser intensities considered are below 5×10^{13} W/cm² such that the ionization and Coulomb explosion processes can be safely ignored.] This novel phenomenon of laser-induced bond-hardening of high-lying VQE resonances is a counterpart of the "bond softening" of low-lying levels as they occur simultaneously in strong fields. The origin of the bond hardening phenomenon can be understood by means of the intensity-dependent avoided crossings of VQE levels and the strengths of non-adiabatic radial couplings. (See also Fig. 11.) We note that population trapping of H_2^+ at 775 nm laser fields has been recently observed experimentally. (c) We performed a nonperturbative study of the dynamical behavior of H_2^+ in intense femtosecond short laser pulse at 775 nm by means of numerical integration of the time-dependent Schrödinger equation using Fourier grid Hamiltonian discretization and the split-operator FFT time propagator. Significant stabilization and trapping of high-lying vibrational states and chemical bond hardening are predicted for both cw lasers and short laser pulses. The wavepackets are seen to be temporarily trapped first at long distances ($\approx 6a_0$) and later on become redistributed to shorter internuclear distances during the turn-off period of the pulses. (See Fig. 12.) (d) Subsequently we performed a theoretical study of the structure of laser-induced VQE resonance states of D_2^+ in two-color laser fields. Stabilization and chemical bond hardening of high-lying VQE resonances are observed. Further it is found that by tuning the relative phase between the fundamental and the third harmonic fields, the electronic-field potential surface and MPD/ATD rates can be modified significantly, suggesting some degree of coherent control of multiphoton dynamics may be possible. (e) More recently, we introduced a procedure for the determination of the kinetic energy spectra of diatomic

molecules undergoing multiphoton above-threshold dissociation (ATD). The method is based on the extension of the non-Hermitian Floquet theory of VQE resonances and an integral equation formulation of the partial rates (branching ratios). The theory is applied to the study of the ATD energy spectra of H_2^+ in 775 nm laser fields.

TABLE I. Partial rates for the detachment by 10.6- μ m radiation.

Number of photons absorbed	Partial rates (a.u.)		
	$I = 10^{10}$ W/cm ²	$I = 5 \times 10^{10}$ W/cm ²	$I = 10^{11}$ W/cm ²
8	0.7117×10^{-9}		
9	0.2034×10^{-9}		
10	0.4323×10^{-10}		
11	0.5258×10^{-11}	0.4074×10^{-6}	0.4321×10^{-5}
12	0.7855×10^{-12}	0.4882×10^{-6}	0.3137×10^{-5}
13	0.2270×10^{-12}	0.3692×10^{-6}	0.2480×10^{-5}
14		0.1295×10^{-6}	0.1237×10^{-5}
15		0.9716×10^{-7}	0.1224×10^{-5}
16		0.8524×10^{-7}	0.1008×10^{-5}
17		0.4881×10^{-7}	0.5046×10^{-6}
18		0.2057×10^{-7}	0.3640×10^{-6}
19		0.7867×10^{-8}	0.4252×10^{-6}
20		0.3944×10^{-8}	0.4275×10^{-6}
21		0.2923×10^{-8}	0.3336×10^{-6}
22		0.2369×10^{-8}	0.2120×10^{-6}
23		0.1765×10^{-8}	0.1169×10^{-6}
24		0.1180×10^{-8}	0.6257×10^{-7}
25		0.7168×10^{-9}	0.3936×10^{-7}
26		0.4017×10^{-9}	0.3262×10^{-7}
27		0.2103×10^{-9}	0.3143×10^{-7}
28		0.1039×10^{-9}	0.3029×10^{-7}
29			0.2747×10^{-7}
30			
31			
32			
33			
34			
Total rate	0.9646×10^{-9}	0.1668×10^{-5}	0.1609×10^{-4}

III. Figure Captions and Figures

- Fig. 1 Comparison of experimental and calculated intensity-averaged multiphoton detachment rates of H^- for linearly polarized Gaussian pulses with peak laboratory-frame intensity of 4 GW/cm^2 . The theoretical results are obtained from perturbative calculations.
- Fig. 2 The frequency- and intensity-dependent complex quasienergies ($E_R, -\Gamma/2$) of H^- for $I = 4, 8, 12, 16$, and 20 GW/cm^2 and $\omega = 0.20 - 0.42 \text{ eV}$; (a) E_R (real energies), showing the ac Stark shifts of H^- ground state, and (b) Γ (imaginary energies), showing the multiphoton detachment rates.
- Fig. 3 Comparison of intensity-averaged multiphoton detachment rates of H^- for the case of laser intensity (in the laboratory frame) $= 4 \text{ GW/cm}^2$. Theoretical prediction . . . , Los Alamos experimental data $\circ \circ \circ \circ$.
- Fig. 4 (a) Electron energy distribution after multiphoton above-threshold detachment of H^- by $10.6 \mu\text{m}$, 10^{10} W/cm^2 laser field. The heights of the bars correspond to the partial rates after absorption of n photons, starting with $n_{\min} = 8$. (b) The same as Fig. 4(a) for the intensity $5 \times 10^{10} \text{ W/cm}^2$, $n_{\min} = 11$. (c) The same as Fig. 4(a) for the intensity 10^{11} W/cm^2 ; $n_{\min} = 16$.
- Fig. 5 Electron angular distributions ($1/\sin\theta$) ($d\Gamma_r/d\theta$) (in units of 10^{-10} a.u.) for the first six above-threshold peaks in the energy spectrum, for the laser intensity 10^{10} W/cm^2 and the wavelength $10.6 \mu\text{m}$. Graphs A–F correspond to the number of absorbed photons $n = 8$ –13, as indicated.
- Fig. 6 Electron energy distributions after multiphoton above-threshold detachment from H^- by $10.6 \mu\text{m}$ radiation and its third harmonic with the intensities $I_L = 10^{10}$ and $I_H = 10^9 \text{ W/cm}^2$, respectively. The heights of the bars correspond to the partial rates after absorption of n fundamental frequency photons, starting with $n_{\min} = 8$. Phase shift (A) $\delta = 0$, (B) $\delta = \pi$, (C) $\delta = \pi/2$, (D) $\delta = -\pi/2$.
- Fig. 7 Electron angular distributions ($1/\sin\theta$) ($d\Gamma/d\theta$) for the nine-photon peak in the energy spectrum. Graphs A–D correspond to detachment of H^- by $10.6 \mu\text{m}$ radiation and its third harmonic with the intensities $I_L = 10^{10}$ and $I_H = 10^9 \text{ W/cm}^2$ and phase shift (A) $\delta = 0$, (B) $\delta = \pi$, (C) $\delta = \pi/2$, (D) $\delta = -\pi/2$.
- Fig. 8 Angle-integrated electron energy spectra dP/dE for multiphoton above-threshold detachment of H^- in a Gaussian pulse with peak intensity $5 \times 10^{10} \text{ W/cm}^2$, and pulse length T . (A) $T = 5 \text{ ps}$, (B) $T = 10 \text{ ps}$, (C) $T = 20 \text{ ps}$, (D) monochromatic results.

- Fig. 9 Potential-energy curves for the ground ($1s\sigma_g$) and first excited ($2p\sigma_u$) states of H_2^+ as a function of internuclear separation R . Also shown is a schematic diagram showing MPD/ATD processes.
- Fig. 10 Multiphoton dissociation (half) widths ($\Gamma/2$) of the vibrational quasienergy resonance states of H_2^+ in the 775-nm laser fields for the laser intensity $I =$ (a) 10^{11} W/cm², (b) 5×10^{12} W/cm², and (c) 5×10^{13} W/cm², showing the rapid reduction of the photodissociation rates of the high-lying resonances with the laser intensity. Here v' labels the order of vibrational quasienergy resonances according to the magnitude of their energies.
- Fig. 11. Vibrational quasi-energy level structure and dressed adiabatic potentials of H_2^+ corresponding to Fig. 10. (a) $I = 10^{11}$ W/cm²; (b) $I = 5 \times 10^{12}$ W/cm², (c) $I = 5 \times 10^{13}$ W/cm². The horizontal line segments at the left side column represent the converged (real parts of the) energies of the vibrational quasi-energy resonances, and the line segments at the right columns represent the energy levels supported merely by the corresponding field-induced adiabatic potential wells. Each adiabatic potential curve is labeled asymptotically by the Floquet channel index $|g \text{ (or } u), n \rangle$.
- Fig. 12. Time evolution of the wavepacket of the $v = 14$ highly excited H_2^+ molecule in a Gaussian laser pulse with pulse length 100 fs and peak laser intensity 5×10^{12} W/cm².

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

IV. Publications Resulting From This DOE Grant Support (DE-FG02-90ER14159)

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